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1 January - 31 December 1987

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ACCELERATOR DEPARTMENT

Annual Progress Report

1 January - 31 December 1987

Abstract. A description is given of research in the fields of chemical reactivity, radiation physics, physical dosimetry and technological application of radiation and irradiation technology, as well as of the operation of various irradiation facilities.

March, 1988

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CONTENTS

Page

PREFACE

1. CHEMICAL REACTIVITY 6

1.1 Decomposition of ozone in aqueous acidic solution 6

1.2 Oxygen isotope exchange in the decay of ozone in acid solution 6

1.3 Light emission from ozone decomposition 7

1.4 Rate constant of $HO_2 + O_2^-$ at elevated temperatures 7

1.5 Activation energy of the reactions $OH + O_2^-$ and $OH + HO_2$ 7

1.6 Reduction of chromate and dichromate in pulse radiolysis ... 8

1.7 Reactivity of H, OH and e_{aq}^- with nicotinic acid: A pulse radiolysis study 8

1.8 Decomposition of perxenate in acid solution 9

1.9 Radiation-induced isotope exchange in water 9

1.10 γ-radiolyse of aqueous solutions of ClO_2 10

1.11 Properties of As(IV) 11

1.12 Pulse radiolytic study of aqueous cyclo-methionylmethionine. 11

1.13 Formation and stability of intramolecular three electron-bonded species in one-electron-oxidized methionine containing oligopeptides 12

1.14 Kinetic and activation parameters for the intramolecular electron transfer processes in model peptides containing redox pair MeT/Trp and Met/Tyr 13

1.15 Activation energy of the intramolecular electron transfer between tryptophane and tyrosine residues in hen egg-white lysozyme 13

1.16 Intramolecular charge transfer between tryptophane and tyrosine in peptides with bridging prolines 13

1.17 Radiolytic products in water 14

1.18 Contract work for Studsvik Energiteknik AB 14

1.19 References 14

Page

2. RADIATION PHYSICS, DOSIMETRY, AND RADIATION PROCESSING 15

2.1 Particle track structure 15

2.2 Investigation of the alanine dosimeter 17

2.3 Radiation processing 19

2.4 Dosimetry for radiation processing 20

2.5 References 22

3. OPERATION AND MAINTENANCE OF THE IRRADIATION FACILITIES 23

3.1 HRC electron linear accelerator 23

3.2 Febetron, field-emission accelerator 24

3.3 ICT, low-energy accelerator 24

3.4 The ^{60}Co-facilities 24

3.5 Lambda Physik Excimer laser 25

3.6 Flash-photolysis set-up 25

3.7 Chemical luminescence set-up 26

4. EDUCATIONAL ACTIVITIES AND PUBLICATIONS 26

4.1 Lectures 26

4.2 Publications 28

5. IRRADIATION FACILITIES AT THE ACCELERATOR DEPARTMENT 31

6. VISITING SCIENTISTS 33

7. STAFF OF THE ACCELERATOR DEPARTMENT 34

1 Figure.

PREFACE

The Accelerator Department conducts research in chemical reaction kinetics, dosimetry, and in processes based on ionizing radiation contributing to its industrial utilization.

At our disposal are three electron accelerators of 10, 2, and 0.4 MeV, cobalt-60 facilities of 3 and 10 kCi, and a Multigas Excimer Laser equipped with various experimental facilities.

A major research effort is directed towards understanding basic chemical processes better, in particular reaction kinetics. The applied techniques include pulse- and steady-state radiolysis, laser flash-photolysis, stop-flow experiments, and computer simulations.

Research is conducted into the physico-chemical response mechanisms following very densely ionizing radiation and into correlation of radical recombination kinetics with ionization density and rate of energy deposition. Development and application techniques of dosimeters take place for photon, electron, and heavy charged particle irradiations.

Commercial test irradiations are carried out at two of the electron accelerators and at both cobalt facilities, but only the 10-MeV linear electron accelerator is equipped for full-scale industrial purposes. Consultive and advisory assistance is rendered in connection with commercial irradiation, including, e.g. calibration of a customer's dosimeter systems.

This report describes the principal activities in these fields for the period 1 January - 31 December 1987.

The contributions marked * are abstracts of published papers.

1. CHEMICAL REACTIVITY

1.1. Decomposition of ozone in aqueous acidic solution

(K. Sehested, H. Corfitzen, E. Bjergbakke, J. Holcman and E.J. Hart (Port Angeles, WA, USA))

The thermal decomposition of ozone in acid solution is a slow process, which depends on pH, concentration of ozone and oxygen, and temperature. Experiments and computer simulations have shown that the process is a radical chain reaction with OH and HO_2 as the propagating radicals. The termination reaction is the radical-radical reaction, which makes the chain length very large. The radical concentration can be calculated to be of the order of 10^{-12}-10^{-15} mol dm^{-1}, which makes the system very sensitive to impurities. Experiments with 10^{-3}-10^{-2} acetic acid lower the initial decay constant by factors of 5-50 depending on the initial ozone concentration. Later on the decomposition process may eventually accelerate, probably because of the products, hydrogen peroxide, glyoxylic acid and formaldehyde reacting with ozone creating new chains for ozone destruction. It is also concluded that the initiation step responsible for the decomposition in alkaline solution, the reaction of ozone with hydroxyl ions, cannot be the predominant initiation step as it is too slow in acid solution. The acetic acid experiments indicate that the radical chain reaction is not the only one responsible for the decomposition of ozone in acid solution.

1.2 Oxygen isotope exchange in the decay of ozone in acid solution

(K. Sehested, J. Holcman, E.J. Hart (Port Angeles, WA, USA) and Ch.-H. Fischer (Hahn-Meitner-Institut, W. Germany))

The oxygen isotope exchange study of mixtures of $^{18},^{18}O_2$ and $^{16},^{16},^{16}O_3$ in aqueous acidic solutions were continued at the Hahn-Meitner-Institut in Berlin. The composition of the isotopic distribution in the oxygen developed from the ozone decomposition was measured by separating the gas from the liquid in a Van Slyke pipette, measuring the oxygen on a gas chromatograph and the isotopic distribution of $^{32}O_2$, $^{34}O_2$ and $^{36}O_2$ on a mass spectrograph. The results show an isotopic exchange during ozone decomposition of a few percent at room temperature, eventually increasing with temperature. They also indicate a pH dependency with less exchange at higher pH. Further, the

ozone concentration seems to influence the degree of exchange. The measured loss in $^{36}O_2$ indicates that the accuracy of the determination of that species is too low when the $^{34}O_2$ was measured fairly accurately. The exchange with visible light is about 100% at pH 2, but lower at higher pH values.

1.3 Light emission from ozone decomposition

(K. Sehested, H. Corfitzen, J. Fenger and E.J. Hart (Port Angeles, WA, USA))

Ozone is a highly energetic molecule that could be expected to emit light by decomposition into ordinary oxygen. Light emission is also reported, and we tried to monitor the light in the region 300-800 nm with a cooled photomultiplier EMI 9824B by flowing the acid ozone solution through a pyrex glass cell on top of the photomultiplier. The emitted light was defused with a peak around 500 nm. This light was enhanced tremendously using tap water instead of distilled water. We also found that the plastic tubings in the flow-system gave rise to the emitted light. No light could be detected even with 5 mM ozone on filling the cell with the solution using only baked glassware and triply distilled water. The conclusion is that the emitted light originates from organic impurities reacting with ozone. Ozone itself may though emit light at higher wavelengths (1100-1200 nm) corresponding to O_2 ($^1\Delta g$), which is not detectable in our system.

1.4 Rate constant of HO_2 + O_2^- at elevated temperatures

(H. Christensen (Studsvik Energiteknik AB, Nyköping, Sweden) and K. Sehested)

The rate of disappearance of the radicals HO_2 and O_2^- in a bimolecular reaction was measured as a function of pH in the range 1.5-10. The plot of k_{obs} changes character at temperatures above 150°C and becomes very pronounced at 200-300°C. The curves indicate a new pK value at lower pH (3-4), which may be attributed to either a new species or a change in the reaction mechanism.

1.5 Activation energy of the reactions OH + O_2^- and OH + HO_2

(H. Christensen, Studsvik Energiteknik AB, Nyköping, Sweden), K. Sehested and E. Bjergbakke)

A study has been initiated of the elementary reactions OH + O_2^- and OH + HO_2

at elevated temperatures. The rate constants are determined in competition with the reaction OH + H_2. The amount of O_2^- and HO_2 formed is measured at five to six different pressures of hydrogen at each temperature, and the results are evaluated by a computer program that calculates the various rates. The preliminary results indicate that another reaction mechanism may take place at temperatures above 150°C.

1.6 Reduction of chromate and dichromate in pulse radiolysis

(P. Sharpe (National Physical Laboratory, Teddington, England) and K. Sehested)

The dichromate dosimeter is widely used for determination of high doses in industrial radiation processing. Therefore a study of the reduction mechanism of chromate (CrVI) was undertaken. The first step, reduction of Cr(VI) to Cr(V) by H radicals can be observed at the absorption at 360 nm. A reoxidation by OH radicals as well as a disproportionation of Cr(V) to Cr(VI) and Cr(III) is monitored. The OH radical reacts very slowly with Cr(III). Therefore silver ions are introduced in the system to pick up the OH radicals forming $Ag(OH)^+$, which reacts by oxidation of the various reduced forms of chromium. $Ag(OH)^+$ can also be followed optically.

As oxygen does not affect the response of the dosimeter, the HO_2 radical was studied. HO_2 does not react with Cr(VI); therefore the mechanism changes when oxygen is present from the beginning or when it develops as a result of the chromium reduction. The HO_2 can be scavenged by either OH or silver radicals, or it disappears in a bimolecular reaction forming hydrogen peroxide, which in turn will reduce Cr(VI). Thereby, the overall reaction will be the same with and without oxygen. The Cr(IV) species is unobserved, which may indicate a fast disproportionation to Cr(III) and Cr(V). The reaction of H_2O_2 with Cr(VI) was studied at longer wavelengths and at longer time scales.

1.7 Reactivity of H, OH and e^-_{aq} with nicotinic acid: A pulse radiolysis study

(S. Solar (Institut für Theoretische Chemie und Strahlenchemie der Universität Wien, Austria), W. Solar (Ludwig Bolzman Institut für Strahlenchemie Wien, Austria), N. Getoff (Institut für Theoretische Chemie und Strahlenchemie der Universität Wien, Austria), J. Holcman and K. Sehested)

The reactivity of aqueous nicotinic acid (NA) towards OH, e^-_{aq} and H-atoms has been investigated in the pH-range 0.3 - 13.8. The OH attack on NA ($k = (2.5 \pm 0.2)\times10^9$ $M^{-1}s^{-1}$) and its N-protonated forms ($k = (2.2 \pm 0.2)\times10^7$ $M^{-1}s^{-1}$) gives OH-adducts with pH-dependent optical spectra: $N\mathring{C}_5H_4(OH)COO^-$: $\lambda_{max} = 310$ nm, $\varepsilon_{310} = 1800 \pm 200$ $M^{-1}cm^{-1}$ and $^+HN\mathring{C}_5H_4(OH)COO^-$: $\lambda_{max} = 325$ nm, $\varepsilon_{325} = 2000 \pm 200$ $M^{-1}cm^{-1}$. $pK_a = 4.5 \pm 0.2$ was determined for these two transients.

The reaction of H-atoms with NA in the pH range 6 - 12 ($k = (6.0 \pm 0.5)\times10^8$ $M^{-1}s^{-1}$ results in the formation of one type of transient, the H-adducts on ring carbons ($\lambda_{max} = 315$, $\varepsilon_{315} = 4500 \pm 200$ $M^{-1}s^{-1}$; pK = 6.7 ± 0.2). With the N-protonated forms of NA, however, two kinds of radicals are produced: pyridinyl ($\lambda_{max} = 285$ nm, $\varepsilon_{285} = 7800 \pm 200$ $M^{-1}cm^{-1}$; $\lambda_{max} = 340$ nm, $\varepsilon_{340} = 4600 \pm 200$ $M^{-1}cm^{-1}$).

The reaction of e^-_{aq} with NA was reinvestigated. The kinetic and spectroscopic data are in good agreement with those previously reported.

1.8 Decomposition of perxenate in acid solution

(U.K. Kläning (Institute of Chemistry, University of Aarhus), K. Sehested and E. Bjergbakke)

The kinetics of decomposition in aqueous perchloric acid of perxenate to xenon trioxide and oxygen was studied by the stopped-flow technique. The decomposition of perxenic acid in strongly acid solution is a first-order process. At pH > 1.5 the decomposition is autocatalyzed by xenon trioxide. At low acidity and perxenic acid concentration kinetic measurements were not reproducible. A mechanism derived from pulse radiolysis studies and in which the first step consists of a formation of hydroxyl radical explains these observations. The formation of hydroxyl radicals is shown to be thermodynamically feasible.

1.9 Radiation-induced isotope exchange in water

(U.K. Kläning (Institute of Chemistry, University of Aarhus), K. Sehested, E. Bjergbakke and E. Larsen (Chemistry Dept.))

^{18}O-enriched water containing N_2O or BrO_4^- with ^{18}O natural abundance were γ- and electron pulse-irradiated. The isotopic composition of oxygen evolved

by the irradiation was determined mass spectrometrically. In alkaline solution (at pH > 11.5) the fraction of ^{18}O in the evolved oxygen is the same as that of ^{18}O in the solvent water. In neutral and acid solution the isotopic composition of O_2 is between that of the dissolved N_2O or BrO_3^- and that of the solvent water. Furthermore, the isotopic composition is constant, independent of pH, dose rate, and whether the solution contained N_2O or BrO_3^-. The results obtained in alkaline solution suggest that the exchange reaction

$$OH + {}^{18}OH^- \rightarrow OH^- + {}^{18}OH$$

is fast.

The observation in acid and neutral solution may suggest that the observed exchange takes place as a cage reaction:

$$O^- + H_2{}^{18}O \rightarrow OH + {}^{18}OH^- \rightarrow {}^{18}OH + OH^-$$

in competition with a process by which $^{18}OH^-$ and OH diffuse from one another.

1.10 γ-radiolysis of aqueous solutions of ClO_2

(U.K. Kläning (Institute of Chemistry, University of Aarhus), K. Sehested and E. Bjergbakke)

G-values for formation of O_2, $ClO^-/HClO$, ClO_2^- and ClO_3^- in O_2 Ar-saturated and N_2O-saturated ClO_2 solutions were measured at pH 5 to 13. Yields of O_2 were measured gaschromatographically by use of a van Slyke apparatus. $ClO^-/HClO$ and ClO_2^- were determined iodometrically. ClO_3^- was determined by reduction with Fe^{2+} to Cl^- in half-concentrated sulfuric acid. Previously the rate constant for the disappearance of ClO_2 by reaction with O^- and with OH was measured. The present measurements show that the reaction of ClO_2 with OH and with O^- proceeds via the channels

$ClO_2 + OH$	$HClO + O_2$	$k = 1.4\times10^9\ M^{-1}s^{-1}$
	$ClO_3^- + H^+$	$k = 2.6\cdot10^9\ M^{-1}s^{-1}$
$ClO_2 + O^-$	$ClO^- + O_2$	$k = 5.4\times10^8\ M^{-1}s^{-1}$
	ClO_3^-	$k = 2.2\times10^8\ M^{-1}s^{-1}$

1.11 Properties of As(IV)

(U.K. Kläning (Institute of Chemistry, University of Aarhus), K. Sehested and B.H.J. Bielski (Brookhaven National Laboratory, New York, USA))

As(IV) was studied by pulse radiolysis of aqueous solutions of As(III) and As(V). Two forms of As(IV) are observed by the reaction of OH/O^- with arsenious acid and arsenite. In neutral and dilute alkaline solution the primary formed As(IV) species is converted to another As(IV) species. In acid solution an equilibrium between the two forms may be observed. In strongly alkaline solution and by reduction of arsenate by the hydrated electron only the secondary formed As(IV) species is observed. The secondary formed As(IV) species disappears in second-order reactions.

Both forms of As(IV) react with O_2 under formation of O_2^-/HO_2, the secondary formed As(IV), however, via a complex with O_2. The following reactions and equilibria are observed:

$$H_2AsO_4^- + e_{aq}^- + H^+ \rightarrow HAsO_3^- + H_2O \qquad k = 2\times10^9\ M^{-1}s^{-1}$$

$$HAsO_4^{2-} + e_{aq}^- \rightarrow AsO_3^{2-} + OH^- \qquad k = 2\times10^6\ M^{-1}s^{-1}$$

$$As(OH)_2O^- + O^- \rightarrow AsO_3^{2-} + H_2O \qquad k = 2\times10^9\ M^{-1}s^{-1}$$

$$As(OH)_3 + OH \rightarrow As(OH)_4 \qquad k = 6\times10^9\ M^{-1}s^{-1}$$

$$As(OH)_4 \rightleftarrows As(OH)_3O^- + H^+ \qquad pK = 7.3$$

$$As(OH)_4 \rightleftarrows HAsO_3^- + H^+ + H_2O \qquad pK = 3.6$$

$$As(OH)_3O^- \rightarrow HAsO_3^-/AsO_3^{2-} + H_2O$$

$$HAsO_3^- \rightleftarrows AsO_3^{2-} + H^+ \qquad pK = 7.6$$

1.12 Pulse radiolytic study of aqueous cyclo-methionylmethionine

(J. Holcman and K. Bobrowski (Institute of Biochemistry and Biophysics, Polish Academy of Sciences, Warsaw, Poland))

Cyclo-methionylmethionine is the simplest peptide bond containing methionine

derivative where the three-electron-bonded S∴S$^+$ radical cation does not experience an influence from the amino- and carboxy terminals, which play an important role in the chemistry of this type of radical cation in small oligopeptides. Because of the presence of the two peptide bonds it is expected that properties of the S∴S bonded radical cation of cyclo-methionylmethionine are close to those in larger polypeptides and proteins. It is found that the OH-adduct to the S atom absorbing at 390 nm is substantially more stable than those in methionine and small methionine peptides. The OH-adduct is in a pseudobasic equilibrium with S∴S$^+$ radical cation absorbing at 510 nm. It is also found that the S∴S bonded radical cation of cyclo-methionylmethionine is substantially more stable than is observed for similar peptides. Further experiments are in progress.

1.13 Formation and stability of intramolecular three electron-bonded species in one-electron-oxidized methionine containing oligopeptides

(K. Bobrowski (Institute of Biochemistry and Biophysics, Polish Academy of Sciences, Warsaw, Poland) and J. Holcman)

The purpose of this work was to explain complex kinetic behaviour which was observed during oxidation of methionine containing digopeptides (Met-Gly-Met, Met-Ala-Met, Met-Met-Met and Met-Gly-Met-Met). The system was studied in detail at 1.0 < pH < 8.0. It was found that 390 nm absorption must be kinetically associated with the immediate precursor of the 490 nm transient. The kinetic parameters of the two processes were identical. Furthermore, the decay at 390 nm and the growth at 490 nm increases at the same rate as with hydrogen concentration. This suggests a protonation process which should most likely occur at the carboxyl group. Pulse irradiation of the same solution but at neutral pH led to the fast rise in optical density at both wavelengths; this was followed by a decay and secondary increase at 490 nm and 390 nm, respectively. These last results were discussed in terms of the transformation of the S∴S-bonded species. Another possible protonation in three-electron-bonded radicals is a protonation of the S∴N bond, which would immediately result in stabilization of the intramolecular S∴S-bonded radical cation. This process, however, can be excluded since pulse radiolysis of an N_2O-saturated aqueous solution of L-Met-L-Met-L-Ala peptide has not yielded complex kinetic behaviour. The results presented and discussed so far have strongly indicated the concept of the pH-dependent equilibrium: S∴S + H^+ ⇕.

1.14 Kinetic and activation parameters for the intramolecular electron transfer processes in model peptides containing redox pairs Met/Trp and Met/Tyr

(K. Bobrowski and K.L. Wierzchowski (Institute of Biochemistry and Biophysics, Polish Academy of Sciences, Warsaw, Poland) and J. Holcman)

Intramolecular electron transfer in model peptides (Trp-Ala-Val-Gly-His-Leu-Met, Tyr-Gly-Gly-Phe-Met, Tyr-D-Ala-Gly-Phe-Met and Tyr-Phe-Met-Arg-Phe-NH_2•2AcOH) has been studied by pulse radiolysis. The selective formation of the Met[S∴Br] species at the methionine residue was followed by electron transfer from the tryptophane or tyrosine residue. As the tryptophyl/tyrosyl radicals are formed very fast, it was difficult to determine the exact value for the rate constant but rather the lowest limit ($k \geq 10^5\ s^{-1}$). However, in the [Met]-enkephalin, inserting D-Ala instead of Gly has resulted in a significant drop of the rate constant to $4.9 \times 10^4\ s^{-1}$. Activation parameters for the last process are measured. Further experiments are in progress.

1.15 Activation energy of the intramolecular electron transfer between tryptophane and tyrosine residues in hen egg-white lysozyme

(K. Bobrowski and K.L. Wierzchowski (Institute of Biochemistry and Biophysics, Polish Academy of Sciences, Warsaw, Poland) and J. Holcman)

Hen egg-white lysozyme is known to undergo a specific temperature-dependent conformational transition occurring when the temperature is increased from 23° to 38°C. Our preliminary experiments using intramolecular electron transfer between tryptophane and tyrosine residues as a probe for structural rearrangements in the protein have shown that this process is characterized by two different activation energies. The break-point in the Arrhenius-type plot occurs around the 30°-40°C range consistent with conformational changes in lysozyme. Further experiments are in progress.

1.16 Intramolecular charge transfer between tryptophane and tyrosine in peptides with bridging prolines*

(K. Bobrowski, K.L. Wierzchowski (Institute of Biochemistry and Biophysics, Polish Academy of Sciences, Warsaw, Poland), J. Holcman, and M. Ciurak (University of Gdańsk, Institute of Chemistry, Gdańsk, Poland))

A series of TrpH-$(Pro)_n$-TyrOH (n=0,1,2,3) peptides in aqueous solutions was

oxidized selectively at the indole group with pulse-radiolytically generated azide radical, $N_3^{\bullet}$. The rates of intramolecular electron transfer from tyrosine to tryptophyl radical were determined over the temperature range 278-338 K. More than a 50-fold decrease in rate is seen (7.7×10^{6} to 1.5×10^{5} s^{-1}) when going from n=0 to n=3, while the activation energy remains in the range 17 kJ mol^{-1} - 23 kJ mol^{-1}.

1.17 Radiolytic products in water

(E. Bjergbakke, K. Sehested, Z.D. Draganić and I.G. Draganić (Boris Kidrić Institute of Nuclear Sciences, Beograd, Yugoslavia))

The radiolytic products in different natural waters and in selected aqueous solutions were studied by computer simulations based on the standard model of water radiolysis and experimental data on free radicals. The results obtained are relevant to the use of water as solvent in radiochemistry, nuclear chemistry and technology, and radioactive waste disposal.

1.18 Contract work for Studsvik Energiteknik AB

(H. Christensen (Studsvik Energiteknik AB, Nyköping, Sweden) E. Bjergbakke, and O. Lang Rasmussen (Computer Installation))

The work consists of simulation of radiation chemical reactions and mechanisms associated with radioactive waste disposal and reactor chemistry.

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2. RADIATION PHYSICS, DOSIMETRY, AND RADIATION PROCESSING

2.1 Particle track structure

(J.W. Hansen, K.J. Olsen (Dept. of Radiophysics, University Hospital of Copenhagen, Herlev), and M.P. Waligorski (Institute of Nuclear Physics, Krakow, Poland))

Track structure theory. The radiation properties of the amino acid L-α-alanine has been investigated in respect to the formation and recombination of radiation-induced free radicals at very high-ionization densities. The rate of recombination of the $CH_3\dot{C}HCOOH$ radical in irradiated alanine depends on the ionization density and is less than 1% per year for low-LET exposures (LET = Linear Energy Transfer) to doses below 10^4 Gy. At exposures to a high ionization density, e.g. $5 \cdot 10^5$ Gy, the recombination amounts to approximately 13% in 2000 hours after the irradiation, after which stabilized conditions are obtained. These results from low-LET exposures indicate that the recombination is consistent with the assumption of a minimum distance between adjacent stable radicals. This provides a probe for studying the strongly inhomogeneous distribution of energy in the track of heavy charged particles for which increasing recombination rates at low average ionization density, single tracks, have been observed for increasing LET.

Thus a high-ionization density leads to a high concentration of trapped radicals which in organic solids, as is observed, cannot stabilize closer together than approximately 10 nm. Excess radicals formed either by high doses of low-LET radiations of high dose rate or in the track core of high-LET particles, will recombine until an equilibrium is obtained. At a dose of

$5 \cdot 10^5$ Gy an average distance between neighbouring radicals is theoretically found to be 3 nm, which is a factor of 3 below the distance considered to be the minimum for stabilized conditions. Saturation of radical formation and recombination effects are observed. The formation of excess radicals and subsequent recombination after high-LET exposures must occur and is referred to the high doses inhomogeneously deposited in the track of a heavy charged particle and is independent of average dose to the sample.

We have tried to correlate measured radical recombination or decay for fast electron exposures to saturation doses with calculations on energy-density distributions in the track of heavy charged particles in order to predict radical decay after high-LET exposures. We calculated the fraction of energy loss in the track deposited at doses above $5 \cdot 10^5$ Gy and converted this fraction of energy into percentage radical decay by multiplication with the amount of decay observed after fast electron exposures to $5 \cdot 10^5$ Gy. We see a good agreement between experimental data and results obtained from this model (Table 1).

Table 1. Decay in 2000 hours after a dose of 10^3 Gy.

MeV/u	Decay meas. %	Decay calc. %	LET_{av} $MeVcm^2g^{-1}$	E_{lim}/E_{tot} %
16 protons *	3.2	3.6	38	30
6 protons	6	4.7	119	39
4 ^{16}O	7.5	7.0	$7.4 \cdot 10^3$	58
15.2 ^{40}Ca	8	6.2	$2 \cdot 10^4$	52
16.5 ^{238}U	6	7.0	$1.1 \cdot 10^5$	58

E_{lim} = track energy deposited above a dose of $5 \cdot 10^5$ Gy.
* penetrating particle.

It is observed from Table 1 that the decay following the ^{16}O-ion exposure is higher than for the exposure with ^{40}Ca-ions even though the average LETs for the stopping particles in the target are comparable. This is, however, due to the higher initial energy of the ^{40}Ca-ions that is followed by a larger fraction of the total ion energy deposited at high particle velocities where the track is wide.

These investigations are related to theoretical considerations on model description for heavy charged particle and neutron interactions with radiation detectors. They concern observed effects that obscure the evaluation of important parameters in the model.

2.2 Investigation of the alanine dose meter

(J.W. Hansen, M. Wille, and K.J. Olsen (Dept. of Radiophysics, University Hospital of Copenhagen, Herlev) and M.P. Waligorski (Institute of Nuclear Physics, Krakow, Poland))

The amino acid L-α-alanine has been investigated for use as a radiation detector in low- and high-LET radiation fields. The decay of the radiation-induced free radicals in alanine and thus the fading of dose meter response following irradiation have been studied. The irradiations concern: 6-20 MeV electrons, 4-18 kV_p x-rays, ^{60}Co γ-rays, 6 and 16 MeV protons, 4 MeV/u ^{16}O-ions, 15.2 MeV/u ^{40}Ca-ions and 16.5 MeV/u ^{238}U-ions. A determination of the special fading properties after high-LET exposures is essential for the proper use of the dose meter both in general high-LET dosimetry and in radiation protection.

The decay rate of the $CH_3\dot{C}HCOOH$ radical in L-α-alanine depends on the deposited energy density and dose rate. Long-term stability of the dose meter response following low-LET exposures has been investigated by repeated measurements of dose meters exposed to 6 and 20 MeV electrons, 4 and 16 MV_p x-rays and ^{60}Co γ-rays. The dose meters exposed to ^{60}Co γ-rays showed a decrease in signal of 1.9% over a period of 4 years in samples given a dose of 100 Gy. Dose meters exposed to pulsed radiation, i.e. linear-accelerator-produced electrons and x-rays, showed a decrease in response of about 6% over 4 years independent of the dose in the dose range of 10-100 Gy. In exposures with a pulsed 10-MeV electron beam to a dose of $5 \cdot 10^5$ Gy, which is the dose for maximum obtainable response, a decay of 13% has been observed after 2000 hours.

For low average doses, single tracks, from high-LET beams of 15.2 MeV/u ^{40}Ca- and 16.5 MeV/u ^{238}U-ions, a decay in response of 8% and 6%, respectively, in 2000 hours has been measured. For high average doses (approx. 10^5 Gy), overlapping tracks, an increase in response of 5% and 8% for ^{40}Ca- and ^{238}U-ions,

respectively, has been observed with a maximum after approximately 100 hours after exposure. Thereafter, a decay of 8% and 6%, respectively, takes place after 2000 hours. These results for single track exposures show approximately the same decay as results obtained for 6-MeV protons (6%) and 4 MeV/u ^{16}O-ions (7.5%), and contradict an expected larger decay following exposure with particles of the same velocity but with a higher LET. We ascribe this smaller decay, however, to the additional exposure from the heavy-ion-induced radioactivity in the sample itself.

Fast-neutron irradiations have been carried out with a radiotherapy beam of neutrons produced by 12.5-MeV deuterons via a Be(d,n)B-reaction in a thick beryllium target at the cyclotron in the Institute of Nuclear Physics, Krakow, Poland. The mean neutron energy was 5.6 MeV and the gamma contamination of the total neutron + gamma dose was 4%. Exposures were measured with an ionization chamber calibrated in terms of dose to tissue, and were converted into absorbed dose in water. In order to evaluate the effectiveness of alanine relative to ^{60}Co γ-rays to pure neutron doses from measurements performed using a mixed neutron beam, we note that the total beam dose consists of 96% neutrons and 4% gamma components. Assuming additivity of the alanine signals, the signal per unit of mixed neutron beam dose is:

$$S(D_T)/D_T = E_n \cdot R_G \cdot 0.96 + R_G \cdot 0.04,$$

where R_G is the signal per unit of gamma-ray dose and E_n the relative effectiveness of "pure" 5.6-MeV neutrons. From this a relative effectiveness E_n = 0.62 ± 0.03 is obtained.

The relative effectiveness of the alanine dose meter has also some energy dependence for low-LET radiation. X-ray exposures have been carried out at the Institute of Nuclear Physics, Krakow, Poland, using a 250-kV_p x-ray radiotherapy unit filtered to give an average x-ray energy of 90 keV. Exposures were measured in terms of roentgens in air and converted into absorbed dose in water, and the response relative to ^{60}Co γ-rays was 0.83 ± 0.04. The energy dependency of alanine is obtained by correcting the measured effectiveness by the differences in energy absorption between water and alanine expressed by the ratio of mass energy absorption coefficients. This leads to a decrease in sensitivity by 0.90 ± 0.04.

An accurate determination of the relative effectiveness of alanine exposed to

15.2 MeV/u ^{40}Ca-ions, 13 MeV/u ^{76}Ge-ions, and 16.5 MeV/u ^{238}U-ions at the UNILAC accelerator at GSI, Darmstadt, W. Germany, has up to now unfortunately not been possible due to unreliable dosimetry on a very unstable accelerator. As a first step in an accurate determination of dose from these ions the range has been measured using stacked 5 µm-thick radiochromic dye film samples. Calculated ranges are adjusted accordingly.

Postal dosimetry comparisons between various radiation therapy centers in Finland, Poland, USA, and Denmark have been carried out using the alanine dose meters. It is possible to compare doses within an accuracy of about 2% at the 95% confidence level.

2.3 Radiation processing

(A. Miller)

Irradiation of various products were carried out at the 10 MeV-linear electron accelerator at the usual level, which is two days a week. The users of this irradiation service include commercial companies, other Risø departments, various institutions and experimentors from this department. The products were irradiated for purposes of sterilization, modification of polymers, decontamination of spices, and modification of semiconductor parameters. Larger amounts of products were irradiated during the beginning of 1987 due to a break-down at a Danish commercial irradiation facility. Under an agreement with this company we carry out irradiations in case of emergencies such as this.

During 1987 we obtained a clearance for test irradiation of various foods. According to Danish rules all irradiation of foodstuffs is forbidden without clearance. The present clearance is for research purposes only and the volumes of products are limited to small amounts. Radiation processing concerns several processes, but it is only the irradiation of food that attracts public attention, although foodstuffs are irradiated on only a very limited scale. We have given informal lectures and newspaper interviews on several occasions, and irradiations have been carried out for students who want to investigate this food conservation technique.

The low-energy accelerator (ICT) and the cobalt facilities have been used for test- and sample irradiations.

Consulting with regard to application of irradiation and industrial processes has been carried out, often in cooperation with the Polymer Group (Chemistry Department).

A total of 35 Danish and 22 foreign customers made use of the irradiation and dosimetric services during 1987.

2.4 Dosimetry for radiation processing

(A. Miller)

Dose validation measurements. Measurement of dose distribution is an essential part of the procedures for approval of an irradiated product, in particular in connection with radiation sterilization and food treatment. We carry out such dose validation measurements for products irradiated at Risø and at Danish commercial irradiation facilities. International rules have not yet been established that specify the detail needed for such measurements, but we take part in the writing of such rules with American organizations (ASTM) and with British organizations (UK Panel on Gamma and Electron Irradiation). In Denmark the rules for sterilized products are being changed from voluntary to obligatory ones. The radiation sterilization rules are under considerations in this connection.

Traceability. We maintain traceability of our dose measurements by intercomparison with National Bureau of Standards (NBS), USA, and National Physical Laboratory (NPL), UK, both for electron- and gamma irradiation. The agreement for gamma measurements are within 1% between Risø and NPL and within 3% between Risø and NBS. Differences of up to 5%, which were reported earlier, have been traced to geometrical differences between the irradiation conditions at the different laboratories. Intercomparisons of dose readings for 10-MeV electron irradiation are still being developed. At present the agreement is better than 1% for dose readings with calorimeters from NBS, NPL and Risø, when calorimeters from all three institutions were irradiated at NPL. These calorimeters are being developed for reference measurements at high-energy electron irradiation.

Dose calibration. We calibrate dosimeters for customers in order to establish calibration data for their dosimetry system. Calibration irradiations were carried out at the cobalt facilities and at the 10-MeV electron accelerator.

In order to verify the established calibration we have sent reference dosimeters to the customers for irradiation at their facility. We use primarily film dosimeters for these purposes, but recently we have also used dichromate dosimeters.

IAEA. We serve as consultants for the development of the International Dose Assurance Service of the IAEA. The service is in operation for gamma facilities all over the world, and the development concerns the use for electron accelerator facilities. Under a technical contract with IAEA (No. 4748), we are developing film dosimeters for use for this purpose. The main problem in connection with the film dosimeters concerns diminishing the influence of external factors such as humidity and temperature during irradiation. The packaging of the dosimeter films in vapour-tight barriers seems to provide the best solution to this problem.

Thin film dosimetry. We use radiochromic dye film dosimeters for reference measurements, dose distribution measurements and as routine dosimeters. We produce our own radiochromic film dosimeters in collaboration with Beiersdorf AG, Hamburg. It is our intention to commercialize this dosimeter in collaboration both with Beiersdorf and with Polymer Physik, Tübingen, who are developing a reader for the dosimeters. The first prototype of this reader is being tested. The collaboration with Beiersdorf AG also concerns a dosimeter for reflected light measurements. This dosimeter is made for CERN, Geneva, with whom we have a contract for developing dosimeters for accelerator component testing.

Dichromate dosimetry. In collaboration with NPL, London, the dichromate dosimeter has been introduced at our department. Dr. Peter Sharpe from NPL has spent three months here investigating the reaction mechanisms of this dosimeter (see elsewhere), and we now use the dichromate dosimeter routinely for reference and routine measurements. We find the dosimeter easy to use, and the response is very reproducible.

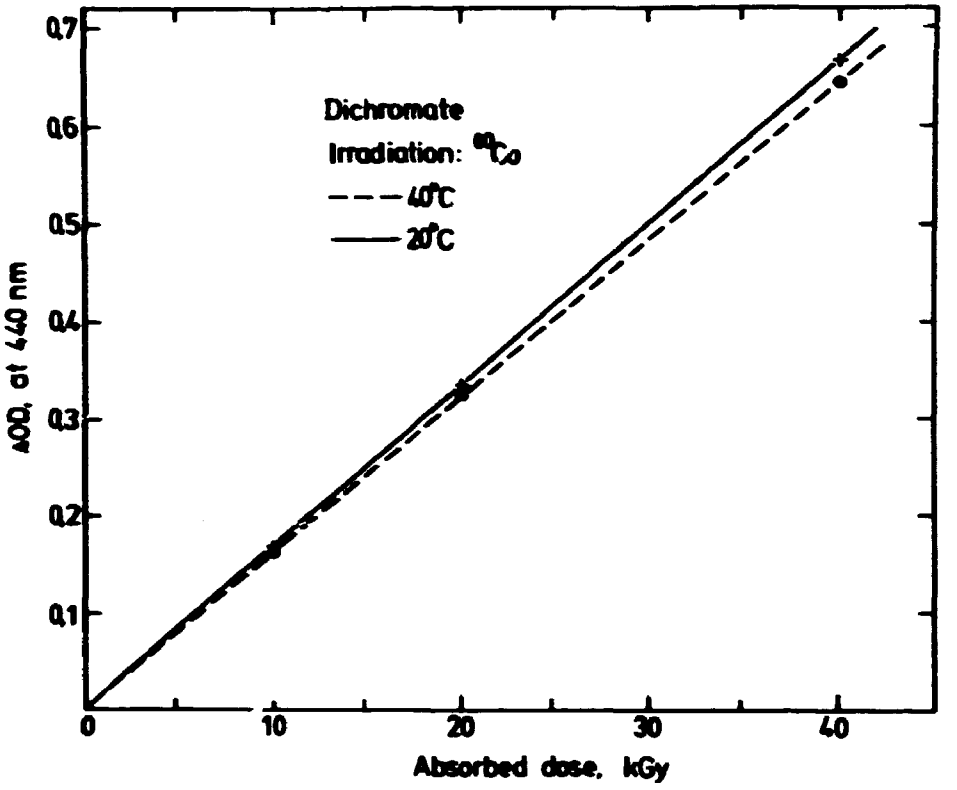

Fig. 1. Calibration curve for the dichromate dosimeter for irradiation at cobalt-60 at the indicated temperatures.

The dosimeter consists of 2 mM $K_2Cr_2O_7$ and 0.5 mM $Ag_2Cr_2O_7$ in 0.1 M $HClO_4$. Water, which has passed through a Millipore filter system, is used. The solution is pre-irradiated to 1 kGy before being filled into 2 ml ampoules, which are flame-sealed. The response function is linear up to 40 kGy, and the lowest dose that can be measured reproducibly (< ± 1%) is approximately 5 kGy.

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3. OPERATION AND MAINTENANCE OF THE IRRADIATION FACILITIES

3.1 HRC electron linear accelerator

(J. Fenger and B. Lynggård)

The accelerator has been in operation for a normal number of hours. About 40% of the time the accelerator was used for pulse radiolysis, 42% for commercial irradiation, 3% for service irradiation and 15% for installation of new components and repair.

A microwave coupler and a klystron (warranty) were sent for repair in the USA. A spare vacuum pump power supply was purchased.

Within the year the following repairs and maintenance were carried out:

Injector system. The high-voltage power supply 250 kV for the electron gun was overhauled after 12 years of operation, 20,000 hours. Some minor components were replaced.

Microwave system. A vacuum leak in the spare klystron (received in 1986) was found by routine check. The klystron was returned to ITT, USA, for a warranty repair.

The main thyratron in the microwave modulator was replaced with one of the two spares after 10,700 hours of operation.

A new trigger was built into the microwave modulator for local operation of the modulator. Local operation of the modulator is used for testing new

klystrons or thyratrons.

Vacuum system. The capacity of the ion pumps in the vacuum system was checked. The efficiency of the vacuum pumps has gradually degraded since its installation in 1975. Steps will be taken to reestablish the performance, either by replacing or regenerating some of the pumps.

Beam handling system. The changes and improvements in the beam handling system include

- a new power supply for the scanning and bending magnets
- a new dose-control system for conveyor irradiation
- a new smoke detector alarm system for product irradiations
- a new beam position detector.

In the water cooling system two pumps malfunctioned, one of them (A12) was replaced, the other, used in the primary cooling system, was repaired. Five flow switches in the primary cooling system were replaced.

3.2 Febetron, field-emission accelerator

The field-emission accelerator was used for pulse radiolysis of gases. The following maintenance was carried out:

A beam tube failed. As we did not have one on stock, parts from two defective tubes were put together, and the result was a tube that has operated successfully now for several months. Nine defective modules were repaired.

3.3 ICT, low-energy accelerator

The low-energy accelerator was used for dosimetry experiments. The pump for the cooling water was replaced.

3.4 ^{60}Co-facilities

10,000 Ci ^{60}Co-facility

The 10,000 Ci ^{60}Co-facility was used for radiation research and for customer

services.

5,000 Ci ^{60}Co-facility

The 5,000 Ci ^{60}Co-cell, presently located in the Control Department of the Danish Serum Institute, Copenhagen, was used for bacteriological research.

3,000 Ci ^{60}Co-facility

The 3,000 Ci ^{60}Co-cell was used for research and test irradiations in radiation chemistry, radiation dosimetry, and customer services. Minor maintenance was carried out.

3.5 Lambda Physik Excimer laser

The laser was used for fast ESR experiments at the Chemistry Department, Aarhus University, and for Raman spectroscopy and flash photolysis at Risø.

The thyratron in the laser malfunctioned and was replaced on warranty. The high-voltage cable, 30 kV, was destroyed by flash-over and was replaced. A leak in the gas circulating system was temporarily repaired; Lambda Physik will supply a modified part.

From time to time there have been problems with the reduction valves in the gas filling system due to exposure to the atmosphere, and a new closed loop system will be designed.

3.6 Flash-photolysis set-up

The system is not yet connected to a computer, thus data collection and handling are difficult. A PC-based system will be designed.

3.7 Chemical luminescence set-up

For measuring very weak chemical luminescence an apparatus, consisting of a cell holder and a sensitive photomultiplier in a cooled house, was designed. Some of the parts were kindly lent from the Health Physics Department.

4. EDUCATIONAL ACTIVITIES AND PUBLICATIONS

4.1 Lectures

K. Bobrowski, J. Holcman, K.L. Wierzchowski, and M. Ciurak, Intramolecular Charge Transfer in Trp-$(Pro)_n$-Tyr (n=0,1,2,3) Studies by the Pulse Radiolysis method. IX Polish Peptide Symposium - "Peptides-87", Sep. 21-25, Putewy, Poland.

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K. Bobrowski and J. Holcman, Three-Electron-Bonded Radical Cations Formation in Pulse Radiolysis of Methionine Peptides. Royal Institute of Technology, Stockholm, Sweden, Nov. 4.

K. Bobrowski and J. Holcman, Formation and Stability of Intramolecular Three Electron Bonds in One Electron Oxidized Simple Methionine Peptides. Hahn-Meitner Institut, W. Berlin, Germany, December.

H. Christensen and K. Sehested, Radiolysis of Aqueous Solutions at Elevated Temperatures. Seminar, Whiteshell Nuclear Research Establishment, Canada, Sep. 9.

J.W. Hansen, Alanine a New Dosimeter for Nuclear Radiation. Seminar at Institute of Occupational Medicine. Lodz, Poland, May 7.

J.W. Hansen, The Alanine Dosimeter as a Test of a Physical Interpretation of Radiation Damage in Solid State Detectors. Seminar at Institute of Nuclear Physics, Krakow, Poland, May 28.

J. Holcman, Reaction Between Hydrogen Peroxide and Superoxide Dismutase. Institute of Applied Radiation, Technical University of Łódź, Poland, June.

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A. Miller, Anvendelse af stråling, tværbinding af plast, sterilisation af hospitalsudstyr, konservering af fødevarer. Risø, 25 March.

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A. Miller, Progress in Calorimetry and Radiochromic Dye Film Dosimetry for Radiation Processing. National University of Mexico, Mexico, June 12.

K.J. Olsen and J.W. Hansen, Radical Recombination in Single Tracks of Heavy Charged Particles. 8th Int. Congress of Radiation Research, Edinburgh, 19-24 July.

K. Sehested and H. Christensen, The Radiation Chemistry of Water and Aqueous Solutions at Elevated Temperatures. 8th Int. Congress of Radiation Research, Edinburgh, 19-24 July.

4.2 Publications

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D. Berenstein, UV-inducible DNA repair in Acinetobacter calcoaceticus. Mut. Res. 183, 219-224.

E. Bjergbakke, S. Navaratnam, and B. Parsons, A quantitative description of the action of high-dose pulses of radiation on aerated acid solutions containing ferrous and chloride ions. Int. J. Radiat. Phys. Chem. Vol. 30, No. 1, pp. 59-62.

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M. Waligórski, J.W. Hansen and E. Byrski, Application of the Alanine Detector to Gamma-Ray, X-ray and Fast Neutron Dosimetry. Institute of Nuclear Physics, Krakow, Raport No 1373/PL.

5. IRRADIATION FACILITIES AT THE ACCELERATOR DEPARTMENT

Electron Accelerators

1. Linear Electron accelerator, Haimson Research Corp.
   Model HRC-712

Specifications:

Electron energy ..... 10 MeV
Average electron current ..... 1 mA
Peak electron current at 10 MeV ..... 1100 mA
Pulse length ..... 0.01 - 4 μs
Pulse repetition rates ..... single pulses and
12.5, 25, 37.5, 50, 100, 150, and 200 pps.

Accelerator room beam facilities:

1. A bent electron beam with scan width of 60 cm in connection with a conveyor system provides a capacity of irradiating, e.g. 400 kg at 35 kGy per hour.
2. Two horizontal beam ports, full average beam power, for electron and X-ray irradiation.
3. One horizontal beam port, reduced average beam power (12.5 pps) in connection with an energy-analyzing spectrometer.

Target room beam facilities:

1. Three horizontal beam ports, reduced average beam power (12.5 pps).

2. Field-emission Electron Accelerator, Febetron Model 705B

Specifications:

Electron energy ..... 1.5 - 2.3 MeV
Peak electron current ..... 4000 A
Pulse length ..... 50 ns

3. Low-energy Electron Accelerator, High Voltage Eng. Corp.
Model EPS 400-IND

Specifications:

Electron energy ..... 400 keV
Electron current ..... 50 mA
Scan width ..... 120 cm
The accelerator is provided with conveyor to permit pilot-plant irradiation.

4. Excimer Laser, Lambda Physik Model EMG102 E[1]

Specifications:

Active medium	F_2	ArF	KrCl	KrF	XeCl	N_2	XeF	
Wavelength	157	193	222	249	308	337	351	nm
Pulse energy (min)	10	200	30	250	150	7	100	mJ
Pulse width (FWHM)		14	9	16	10	6	14	ns
Single pulse and repetition rates up to	80	80	100	100	100	100	50	Hz

^{60}Co-Facilities

10,000 Ci ^{60}Co-facility (built at Risø 1957)
Designed for very homogeneous irradiation of samples with a maximum diameter of 180, 100, or 60 mm. The corresponding maximum dose rates (8.470 Ci (3.15×10^{14} Bq), 1 January 1988) are 5.25×10^{5}, 1.45×10^{6}, and 3.33×10^{6} rad·h^{-1} (1.46, 4.04, 9.2 Gy·s^{-1}), respectively.

5,000 Ci ^{60}Co-facility (built at Risø 1971)
Designed for laboratory use and fitted with a 123 mm$^{\varnothing}\times$150 mm irradiation chamber. The dose rate in the center of the chamber (4.690 Ci (1.75×10^{14} Bq), 1 January 1988) is 3.68×10^{5} rad·h^{-1} (1.02 Gy·s^{-1}). The cell is located at the Control Department, National Serum Institute, Copenhagen.

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[1]An appropriation by the Danish Natural Science Research Council, shared with Aarhus University.

3,000 Ci ^{60}Co-cell (built at Risø 1968)

Designed for laboratory use and fitted with a 120 mm$^{\emptyset}$×220 mm irradiation chamber. The dose rate in the center of the chamber (3.725 Ci (1.38×10^{14} Bq), 1 January 1988) is 3.81×10^{5} rad·h^{-1} (1.06 Gy·s^{-1}).

Sources were replenished in all 3 facilities in April 1986.

6. VISITING SCIENTISTS

K. Bobrowski, Institute of Biochemistry and Biophysics, Warsaw, Poland

H. Christensen, Studsvik Energiteknik AB, Nyköping Sweden.

N. Getoff, Institut für Strahlenchemie, Vienna, Austria.

E.J. Hart, Port Angeles, WA., USA.

U. Kläning, Kemisk Institut, Aarhus Universitet,

W.L. McLaughlin, National Bureau of Standards, Gaithersburg, MD., USA.

P. Sharpe, National Physical Laboratory, Teddington, Middlesex, England.

S. Solar, Institut für Strahlenchemie, Vienna, Austria.

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